Materials Science Forum
ISSN: 1662-9752, Vol. 880, pp 11-14
doi:10.4028/www.scientific.net/MSF.880.11

Submitted: 2016-07-13
Accepted: 2016-07-21
Online: 2016-11-21

Electrospun Polyvinylpyrrolidone (PVP) Nanofiber Mats Loaded by *Garcinia mangostana* L. Extracts

Ida Sriyanti[1,4], Dhewa Edikresnha[1,4], Muhammad Miftahul Munir[2,4], Heni Rachmawati[3] and Khairurrijal[1,4,a]

[1]Physics of Electronic Materials Research Division,

[2]Theoretical High Energy Physics and Instrumentation Research Division, Faculty of Mathematics and Natural Sciences

[3]Pharmaceutics Research Division, School of Pharmacy,

[4]Bioscience and Biotechnology Research Center, Institut Teknologi Bandung, Jalan Ganesa 10, Bandung 40132, Indonesia

[a]krijal@fi.itb.ac.id

Keywords: Polyvinylpyrrolidone (PVP), *Garcinia mangostana* L., Nanofibers.

Abstract. Composite nanofibers of polyvinylpyrrolidone (PVP) and *Garcinia mangostana* L. extract (GME) have been synthesized through electrospinning method for application in drug delivery systems. The precursor solution of 10 mL PVP 10% w/w and GME 2% w/w was then electrospun collected at the rotating collector at the following optimum parameters: a voltage of 15 kV, a collector-nozzle distance of 12 cm, and a flow rate of 1 mL/hour. SEM images showed that the average diameters were 476 nm and 690 nm for the PVP and PVP-GME composite nanofibers, respectively. To some degree, the addition of GME into PVP nanofibers increased the average diameter size of nanofibers. Moreover, the release studies, it was shown that 80% of the GME was released within 30 minutes. Therefore, the PVP-GME composite nanofibers can be applied as the drug delivery systems.

Introduction

Electrospinning is a simple and practical method for the fabrication of ultrafine fibers with an average diameter within the range of sub-micrometer to nanometer by using different types of polymers [1]. The nanofibers can be applied into various fields, particularly in biomedics such as for tissue engineering [2], wound dressings [3] and drug delivery systems (DDS) [4]. For DDS application, electrospinning method can be used to produce drug-loaded nanofibers which can potentially increase the dissolution rate and to control the release of drugs, resulting in an improved bioavailability [5]. Mangosteen (*Garcinia mangostana* L.) is a tropical plant that is cultivated mostly in Southeast Asia, including Indonesia. Mangosteen skin contains active polyphenolic compounds that include xanthones and tannins which have been proven to have pharmacological activities such as antioxidant [6] and anti-cancer [7]. However, the low bioavailability of the compounds has hindered many applications of the drugs [8]. The incorporation of GME into the nanofibers system is expected to increase the biovailability of GME.

Polyvinylpyrrolidone (PVP) was selected because it has been widely used in the health field as the carrier of drug compounds in drug delivery systems [9]. PVP has also received recognition from the FDA (Food and Drug Administration), USA, as a polymer that is safe to be applied in the health industry, especially for medicine and food. Many researchers have successfully produced electrospun PVP nanofibers for a wide range of applications [5,9]. However, to the limit of our knowledge, electrospun composite nanofibers of PVP-GME have not been reported. This paper discusses and evaluates the effect of adding GME into PVP nanofibers regarding about the morphology of the fibers and the release pattern of GME.

Experimental

PVP (molecular weight of 1300 kg/mol; Sigma Aldrich) was used in this study. Ethanol (analytical grade, Merck) was used as the solvent. Mangosteens were purchased from the local market. PVP solution was obtained by dissolving PVP in ethanol at weight concentration of 10% w/w. The PVP solution as a precursor was then electrospun to make PVP nanofibers. To produce PVP-GME nanofibers, another precursor solution was made by mixing the pasta solution of GME with a weight concentration of 2% w/w into the 10% w/w PVP solution and then stirred again at room temperature for ± 2 hours. The precursor solutions were then electrospun at a voltage of 15 kV, a collector-nozzle distance of 12 cm, and a flow rate of 1 mL/hour. The electrospinning process took about 8-10 hours.

Scanning electron microscope (SEM, JEOL JSM-6510LV) was used to characterize the morphology and the diameter of the PVP nanofibers mat and the PVP-GME composite nanofibers mat. Fourier transform infrared spectrometer (FTIR, Bruker Alpha 1-176-396) was used to detect the presence of typical functional groups of the nanofibers mat. The release of GME from the composite nanofibers was characterized using USP Dissolution Apparatus I (Research Hanson SR8 Plus). The release medium used was phosphate buffer and surfactant (1%) at pH of 6.8 with a concentration of 0.1 M. This study was repeated three times at 37°C and 150 rpm. The 5 mL aliquots were taken at 5, 10, 15, 30, 45. 60 and 120 minutes and then replaced with an equal volume of release medium. The amount of GME released was determined by UV-Vis spectrophotometer (Hitachi U-2800) at a wavelength of 320 nm. The viscosity of the solution was measured using Fenske Ostwald Viscometer (Fisher) and the test was repeated three times.

Results and Discussion

After the production of fibers mats, the PVP nanofibers mat and the PVP-GME composite nanofibers mat had different colors. At the macro level, the color of PVP fibers mat was white while the mat of PVP-GME composite nanofibers was yellow which proved that the GME had been evenly mixed into the polymer matrix.

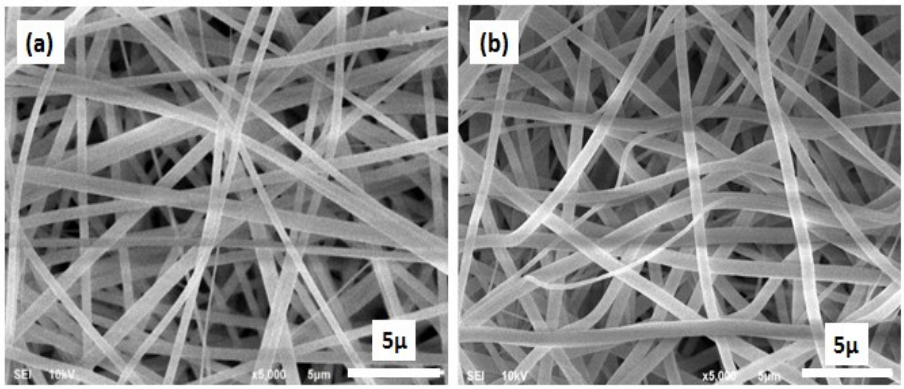

Fig 1. SEM images (5.000×) of (a) PVP nanofibers mat and (b) PVP-GME nanofibers mat.

Figure 1 shows the SEM images of PVP and PVP-GME nanofibers mats. Both nanofibers mats had similar morphology with nanosize average diameters. The addition of GME into the PVP nanofibers did not affect the morphology of the fibers and relatively homogeneous and regular nanofibers were formed. In the previous studies, the addition of emodin into the PVP fibers up until 0.2 % w/v did not hinder the formation of regular fibers [9]. Therefore, one of the factors that determine the formation of the fibers through electrospinning is the concentration of the polymer. The precursor solution used for electrospinning process should have enough polymer concentrations so that the entanglement of polymer chains can form fibers.

The diameter distribution of the nanofibers is shown in Figure 2. For the PVP nanofibers, the measured diameter was distributed within 200-1000 nm with an average of 476 nm and a standard deviation of 5.6 nm. The PVP-GME composite nanofibers mat had a higher average diameter of 690

nm ranging from 200-1800 nm and a standard deviation of 17.29 nm. The viscosity of PVP solution was 112 cP while that of PVP-GME solution was 164 cP. Increasing the viscosity of the solution will increase the number of bonds between the polymer chains that will obstruct the elongation of the fibers during the attraction towards the collector [10]. Therefore, the diameter of the PVP-GME nanofibers mat was greater compared to that of the PVP one. Something similar has been reported by Suwantong, et al. that the addition of GME in the PLA fibers will increase the value of the viscosity and the diameter of the fiber [3].

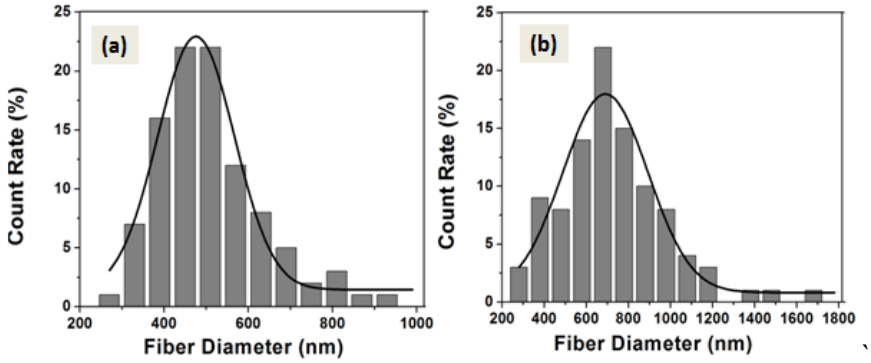

Fig 2.The diameter distributions of (a) PVP nanofibers mat (b) PVP-GME nanofibers mat.

The GME spectrum in Figure 3(a) shows a broad peak at 3326 cm^{-1} which is a hydrogen-bonded group indicating the existence of alcohol and polyphenol compounds in GME [11]. The peak of 2972 cm^{-1} and 2925 cm^{-1} are both identified as asymmetric C-H stretching [12]. The peak at 1639 cm^{-1} is the C=O stretching band [12]. The peak at 1442 cm^{-1} is related to the CH-O-H stretching band [11]. The peak of 524 cm^{-1} is identified as = C-H bending [11].

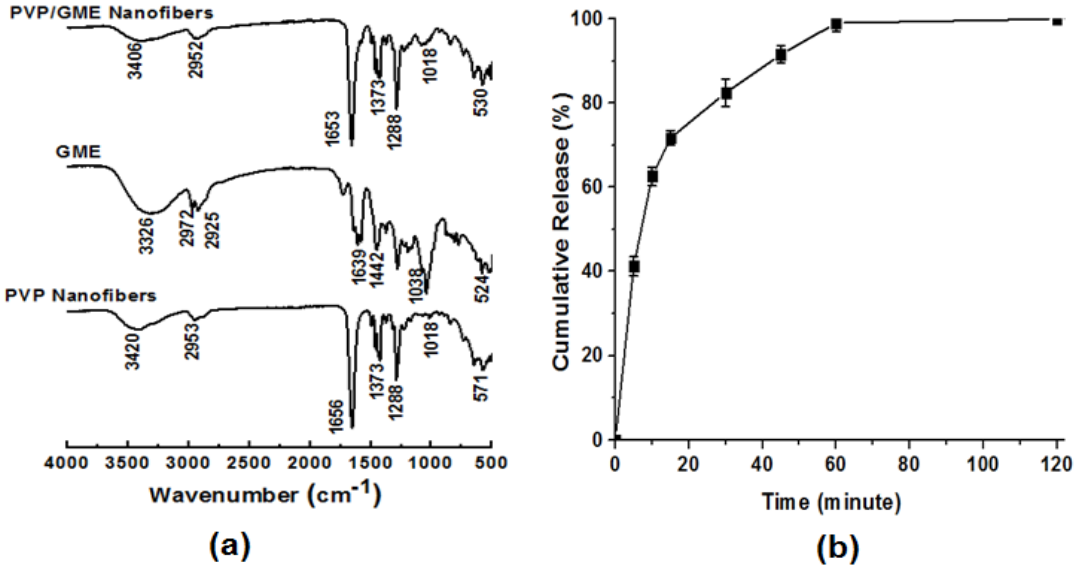

Fig 3. (a) FTIR spectra of PVP nanofibers, GME, and PVP-GME composite nanofibers, and (b) The release of GME from the PVP-GME nanofibers at phosphate buffer solution at pH of 6.8.

Moreover, the PVP nanofibers have peak at 3420 cm^{-1} which indicates O-H stretching band [5,9]. The peaks at 2953 cm^{-1} and 1656 cm^{-1} are the asymmetric stretching of CH_2 and the C-O stretching, respectively [5,9]. The peaks at 1373 and 1288 cm^{-1} are C-H bending and CH_2 wagging, respectively [5]. The peaks at 1018 and 571 cm^{-1} are identified as the CH_2 rock and N-C=O bending [5]. The addition of GME into PVP nanofibers caused shifts of the peaks at 3420, 2953, and 1656 cm^{-1} towards lower wavenumbers, which are 3406, 2952, and 1653 cm^{-1}, respectively, as seen in Figure 3(a).

The release of GME from the PVP-GME composite nanofibers with respect to time can be seen in Figure 3(b). Initially, the release of GME from the PVP-GME nanofibers occurred very rapidly (burst release) such that 80% of GME was released within the first 30 minutes. Afterwards, the release took longer time and finally 100% of the GME was fully released after one hour. Some previous studies also reported the rapid initial release of GME from the CS-EDTA/PVA fibers [11]. The burst release can be caused by the large surface area of the nanofibers, the small diameter size of the fibers and the existence of active substances that was trapped on the surface of the nanofibers during the synthesis process [13].

Conclusion

PVP nanofibers have been synthesized with GME as the active ingredient. The addition of GME into the PVP nanofibers increased the average diameter of the nanofibers compared to pure PVP nanofibers with the same polymer concentration from 476 nm to 690 nm. The addition of GME caused some molecular interactions in the PVP nanofibers indicated by the shift of wavenumber towards smaller wavenumber. The release pattern of GME on the PVP-GME composite nanofibers was burst release. It is therefore concluded that the PVP-GME composite nanofibers can be used for drug delivery systems.

Acknowledgement

This research was financially supported by Directorate of Research and Community Engagement of Ministry of Research, Technology and Higher Education, Republic of Indonesia under the University's Excellent Research (PUPT) Grant in the fiscal years 2015-2016.

References

[1] M.M. Munir, A.B. Suryamas, F. Iskandar and K. Okuyama: Polymer Vol. 50 (2009), p. 4935
[2] H. Yoshimoto, Y.M. Shin, H. Terai and J.P. Vacanti: Biomaterials Vol. 24 (2003), p. 2077
[3] O. Suwantong, P. Pankongadisak, S. Deachathai and P. Supaphol: Polym. Bull. Vol. 71 (2014), p. 925
[4] X. Shen, D. Yu, L. Zhu, C. Branford-White, K. White, N.P. Chatterton: Int. J. Pharm Vol. 408 (2011), p. 200
[5] A. Rahma, M.M. Munir, Khairurrijal, A. Prasetyo, V. Suendo and H. Rachmawati: Biol. Pharm. Bull Vol. 39 (2016), p. 163
[6] N.M. Thong, D.T. Quang, N.H.T. Bui, D.Q. Dao, and P.C. Nam: Chem. Phys. Lett. Vol. 625 (2015), p. 30
[7] Y. Akao, Y. Nakagawa, M. Iinuma and Y. Nozawa: Int. J. Mol. Sci. Vol. 9 (2008), p. 355
[8] A.F.A. Aisha, Z. Ismail, K.M. Abu-Salah and A.M.S.A. Majid: J. Pharm. Sci. Vol. 101 (2012), p. 815
[9] X.Y. Dai, W. Nie, Y.C. Wang, Y. Shen, Y. Li and S.J. Gan: J. Mater Sci: Mater. Med. Vol. 23 (2012), p. 2709
[10] S. Ramakrishna, K. Fujihara, W.E. Teo, T.C Lim and Z. Ma: *An Introduction to Electrospinning and Nanofibers* (World Scientific, Singapore, 2005)
[11] N. Charernsriwilaiwat, T. Rojanarata, T. Ngawhirunpat, M. Sukma and P. Opanasopit: Int. J. Pharm. Vol. 452 (2013), p. 333
[12] M. Ahmad, B.M. Yami and A.M. Lazim: Chem. Cent. J. Vol. 7 (2013), p. 85
[13] C. Risdian, M. Nasir, A. Rahma and H. Rachmawati: J. Nano. Res. Vol. 31 (2015), p. 103